Simulations of Second-Order Non-Linearity Processes

Marco Preuss, University of Applied Science Zwickau, Germany

September 7, 2016

Abstract

Contents

1 Introduction

Synchrotron radiation provides intense, laser-like flux of photons in wide spectral ranges, which is in that way not possible with other sources. Synchrotron radiation sources nowadays show an increasing importance in many areas in science [1]. The first synchrotron source started operating in 1973 [2]. That synchrotron just used bending magnets to produce radiation caused by charged particles moving with relativistic speed. This type of synchrotron sources were the first generation. The so called wigglers and undulators followed. Those are magnets with an alternating polarity, which made higher intensities possible.

The fourth generation of synchrotron radiation sources are free-electron lasers (FEL), such like the Flash at DESY. Those sources provide really wide spectral ranges, reaching from microwaves to X-rays and the achievable brilliance is extremely high. The effect that generates the radiation in FELs is called self-amplified spontaneous emission (SASE). With SASE it is possible to emmit quasi-coherent photons, but due to spontaneous generation from white-noise fluctuations within the electron bunches every single shot is slightly different [3], they jitter. That is to say pulses are different in pulse energy, beam profile and arrival time.

This causes the need of methods that improve the beam quality. A method to do that is seeding the FEL. Those Seeding methods anable higher beam qualities that are important for time-resolved experiments or pump-probe laser systems. If one wants to seed the FEL there are some requirements to the seeding source which are crucial to increase beam quality. Such properties for example are high repitation rates, tuneable wavelengths over the hole spectral range of the FEL or spatial properties. One of the most important aspects is the conversion efficiency when unsing nonlinear effects to generate various wavelengths.

This report to DESY's summer students programm deals exactly with the topic of conversion efficiency in nonlinear processes. Some basic issues that influence the efficiency are discussed in chapter 2 on the example of a second harmonic generation (SHG). Furthermore simulations were done to estimate the best parameters for an experimental setup of a SHG. The last part of this report is about setting up the SHG in the lab and compare the results with the before calculated data.

2 Theoretical Background

2.1 Second order Nonlinearities

The basic principle behind every nonlinear process is that the polarisation, induced in a material, behaves not linear with respect to the electromagnetic field that caused electric dipole moments. In case of linear optics one can discribe the polarisation by all dipole moments in a certain volume:

$$P(\omega) = \varepsilon_0 \chi^1 E(t) \quad (1)$$

But this equation does not hold anymore when lasers with high intensities are used. If the intensities are just high enough non-linear effects accure and equation 1 has to be extended:

$$P(\omega) = \varepsilon_0 [\chi^1 E(t) + \chi^2 E(t) + \chi^3 E(t) + ...] \quad (2)$$

The second order non-linearity χ^2 , must be high when a SHG is intended. χ^3, that is to say, third order non-linearities belong to effects like selfphase modulation and optical Kerr effect. Second order processes are SHG, sum-frequency generation (SFG) and difference-frequency generation (DFG).

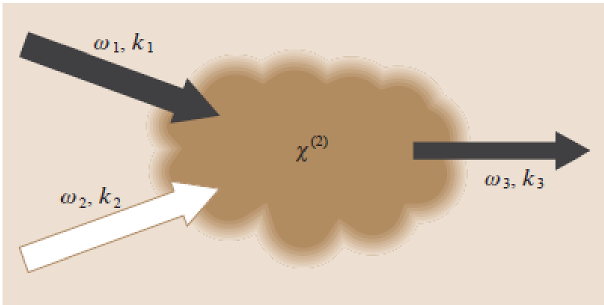


Figure 1: Sum-frequency generation $\omega 1 + \omega 2 = \omega 3$ in a medium with a quadratic nonlinearity. If $\omega 1 = \omega 2$ second-harmonic generation occurs [4]

For the purposes of this report only SHG should be discussed briefly. Assume an electromagnetic wave passes through a crystall, that shows a quadratic non-linear behavior. This wave, with the frequency ω, is called the pump wave. The properties of the non-linear medium cause the pump wave to generate a new wave with the frequency of 2ω. Equation 2 could than look like this:

$$P^2 = 2\varepsilon_0 \chi^2 E E^* + \varepsilon_0 \chi^2 E^2 e^{-i2\omega t} \quad (3)$$

As one can see, the first term does not have a frequency component. This is due to optical rectification [5], a non-linear effect that generates a quasi-DC polarization in a non-linear crystall. That means the first part of Equation 3 stands for a static field. The second term is that of the SHG. It shows an oscillation with 2ω, that is o say, twice the frequency of the incoming pumpwave.

2.2 Phase Matching and Tuning Curves

Figure 2 shows a sketch of a simplified collinear Second-harmonic generation. As mentioned in the chapter before, the fundamental wave (red) induces dipols that oscillate with a frequency two times higher than the fundamental wave. This leads to the second-harmonic, which is showen in Figure 2 by the blue wave.

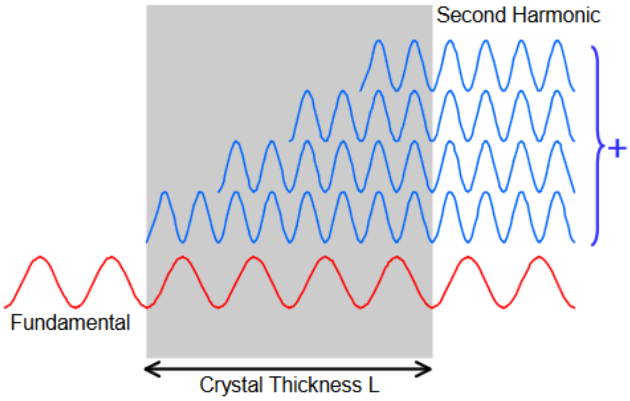


Figure 2: Principle of a phase matched SHG. The picture shows the case where all dipols radiate in phase in the forward direction, so that all contributions add up constructivly [6]

One can then derive the efficiency of the SHG from the relative phase of the dipols. In general the efficieny is the sum of all parts adding up over the crystal lengthn (crystal length is oriented in direction of the z-axis). For that reason, an integral leads to the conversion efficiency:

$$\eta = \frac{1}{L^2}\left|\int_0^L e^{i2k_w z}e^{-i2k_{2w}z}\right|^2 = \frac{1}{L^2}\left|\int_0^L e^{i\Delta kz}\right|^2 = \left(\frac{sin(\Delta kL/2)}{\Delta kL/2}\right)^2 \tag{4}$$

From that equation one can see that the phases result in a difference of the wave vektors. This difference is the wave vector mismatch with $\Delta k = 2k_\omega - k_{2\omega}$. If the mismatch equals zero this function has a maximum. By implification that means an efficient SHG is just possible with a phase matched process.

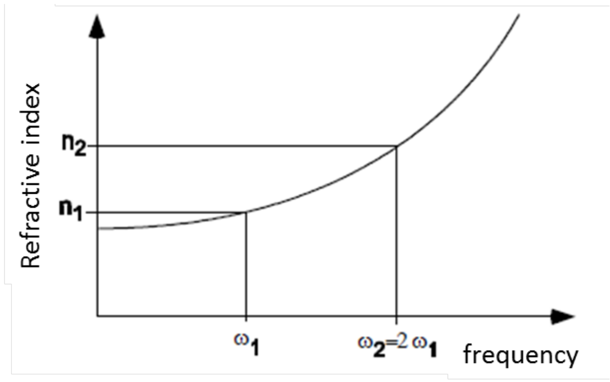


Figure 3: Frequence-dependency of the refractive index for normal dispersion[7]

For the case of normal dispersion, the refractive index increases with increasing frequency. That behavior is showen in figure 3. With the case of a SHG a problem is observed. The phase matching condition ($\Delta k = 0$) leads to the following expressions:

$$\Delta k = 2k_\omega - k_{2\omega} = 0 \tag{5}$$

$$k_{2\omega} = 2k_\omega \tag{6}$$

$$n_{2\omega}\omega_2 = 2n_\omega\omega_1 \tag{7}$$

The phase matching condition showes that the refractive indices $n_{\omega 1}$ and $n_{\omega 2}$ must be the same to fullfill phase matching (with the condition of $\omega_2 = 2\omega_1$). This implies that phase matching is not possible in the boundary conditions of normal dispersion. One technique to avoid this problem is phase matching by birefringent materials like Lithium triborate (LBO) or Beta-Barium borate (BBO).

The principle why phase matching with birefringend crystals work is that different polarisations have different refractive indeces. This fact leads then to matched refractive indeces for ordinary (o) and extra-ordinary (e) polarised parts. In other words, the frequency dependant refractive index curve can now be shiftet so that $n_e(2\omega) = n_o(w)$. This is showen in figure 4.

There are different kinds of phase matching types. One way to distinguish the typ of phase matching is the sort of the included polarizations. If the incident electro magnetic waves are not different in polarisation to each other it is called typ 1 phase matching (e.g. oo→ e). If the fundamental wave possesses ordinary parts as well as extra-ordinary parts then it is typ 2 phase matching (e.g. oe → e).

From crucial importance is than obviously which type of crystall is used. Choosing the right type of crystall is necessary for the successful generation of a SHG or higher-harmonics. The following chapter discusses some aspects of those materials.

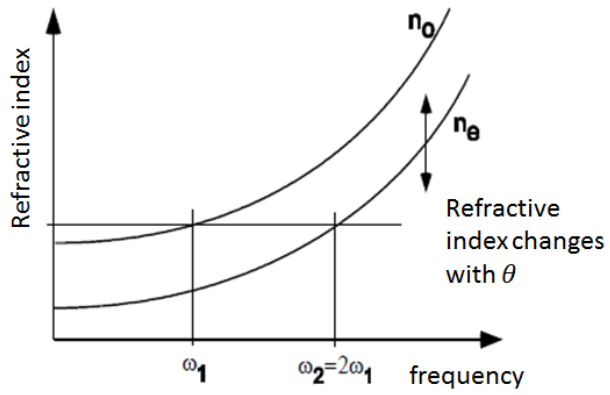


Figure 4: Phase matched refractive indeces for ordinary and extra-ordinary polarization. θ is the tuning angle of the crystal (see following chapter)[7]

2.3 Birefringend Crystals

Another really important characteristic for the manner of phase matching is obviously the type of the used briefringend crystall. One distinguishes non-linear crystalls by it's behaviour they show for the different polarisations. That is to say:

$$negative : n_e < n_o \tag{8}$$

$$positive : n_e > n_o \tag{9}$$

Against this background it is clear that the plot from figure 4 of the last chapter shows phase matching with a negative crystal. Than there are uniaxial (e.g. BBO) and biaxial (e.g. LBO) crystalls. To clarify what that means one should take notice of figure 5. Those are so called index ellipsoides.

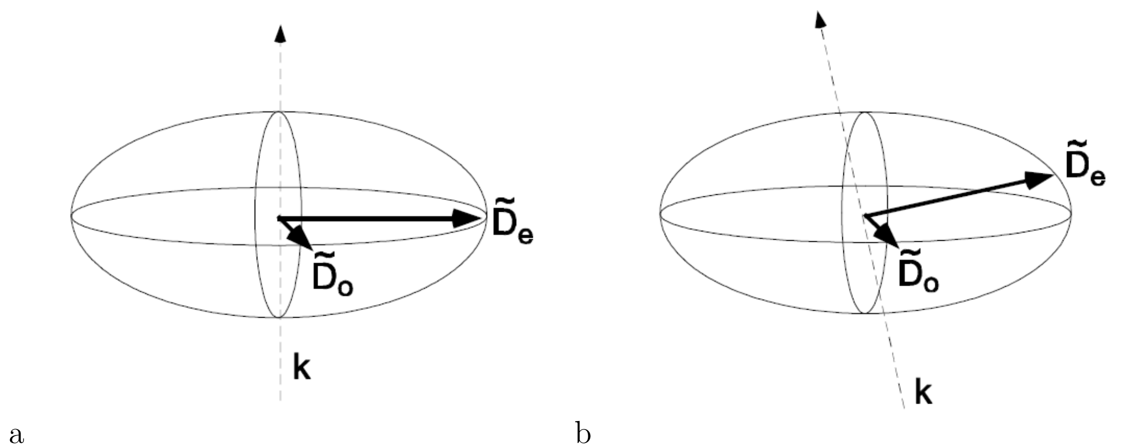


Figure 5: a: index ellipsoide with $\theta = 0°$ **b:** index ellipsoide with $\theta \neq 0°$ [7]

The Vector $\tilde{D}$ is described by the electric displacement field and the energy density (see equ. 10). If one then denotes the energy equation (see equ. 11) to this vector it shows that the length of $\tilde{D}$ is independant of the light's intensity. The lengths of the coordinate axis are than determinded by the refractiv indeces.

$$\tilde{D} = \frac{1}{\sqrt{8\pi W_e}} D \tag{10}$$

$$\frac{\tilde{D}_x^2}{n_y^2} + \frac{\tilde{D}_x^2}{n_y^2} + \frac{\tilde{D}_z^2}{n_z^2} = 1 \tag{11}$$

A common convention is to set the direction of the wave vector in the same direction of that from the optical axis. This leads to the classification of non-linear crystals addressed at the beginning of this chapter. For the case that two refractive indeces are equal, ther is just one optical axis and those crystals are therefore called "uniaxial". If the refractive indeces are different in every direction, that crystal has two optical axis and is than called "biaxial".
Assuming an uniaxial crystal in figure 5 and the ordinary part of the incident light is along the optical axis and independant from rotating the crystall.

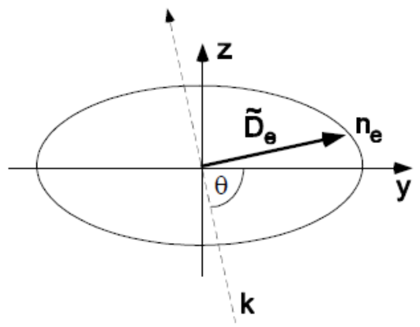


Figure 6: 2-dimensional ellipsoide that shows the dependancy of the refractive index from θ [7]

By rotating the crystall with an angle Θ it is possible to keep the refrective index for ordinary polarized light constant while that of the extra-ordinary has changed (fig. 5b). This is how refrective indeces can be matched by using birefringend crystals. This can be seen in figure 5. Those plots are, if you will, a different way to show the principle of phase matching from figure 4.
Finally for this chapter, there should be a quantitative description of the influence from θ on the refractive indeces. The following equation showes the relation bewtween refractive index and tuning angle.

$$\frac{1}{n_e(\theta)^2} = \frac{sin^2\theta}{n_e^2} + \frac{cos^2\theta}{n_o^2} \tag{12}$$

3 Numerical Simulation

3.1 Introduction to the software

Second-order nonlinearities are important optical processes in optical applications like SHG, optical parametric amplification (OPA) or in optical parametric oscillation (OPO). Because of the fact that the most materials with a second-order non-linear behaviour do also show birefringence, for those processes phase matching by birefringence is choosen as method to match ordinary and extra-ordinary refractive index. Depending on what the exact goal is, the non-linear processes can be very different in crystal material, phase matching angles and so on. One further example for the good efficiency of a non-linear process is the beam diameter. A lot of times it is necessary to focus beams to really small beam waists to get the desired effect. Because of the high intricacy of all these processes it is evident to have computational methods to simulate the interactions. A programm that is able to simulate non-linear processes is Chi2D.
That programm is a software which uses a split-step Fourier method. That works well because most of the second-order non-linear processes are based on that method and can therefore be treated with that kind of numerical method. This so called split-step method treats propagation phenomena in the spectral domain by using fourier transforms. The differential coupled equations are then solved in space-time domain [8]. This software was originally made to study the behaviour of ultra-broadband parametric amplifiers. But above this the numerical simulations include every second-order phenomena and phase matching, as well as walk-off effects and diffraction of the used laser pulses [9]. The next chapter will deal with the interpretation of the simulated data by the example of a Secon -harmonic generation in a LBO-crystal.

3.2 Simulation of a SHG

The main aim of this simulation is to calculate the best parameters for setting up a SHG from $\lambda = 1030nm$ to $\lambda = 515nm$. The very first thing one should eveluate is of course

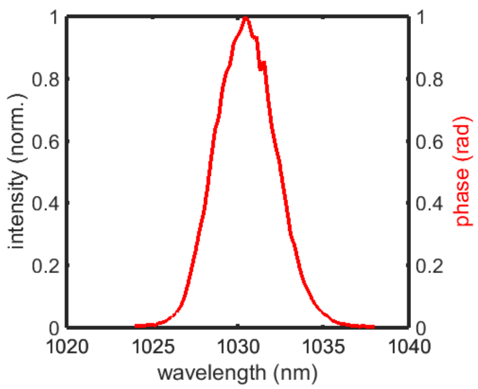


Figure 7: Output spectra from the lasersystem with $\lambda = 1030nm$ and $E = 6\mu J$

the output-spectra from the light source. Therefore the output-spectra was measured and plotted in figure 7. This spectrum was then loaded and used for the simulation. Next step was to choose a non-linear material that fits the SHG. Therefore on needs a crystal that supports the used wave lengths. That is to say a material is needed which is transparent over the spectral range from $1030nm$ to $515nm$. For this purpose Lithium triborate (LBO) was choosen. Its transmittance behaviour can be seen in the following figure.

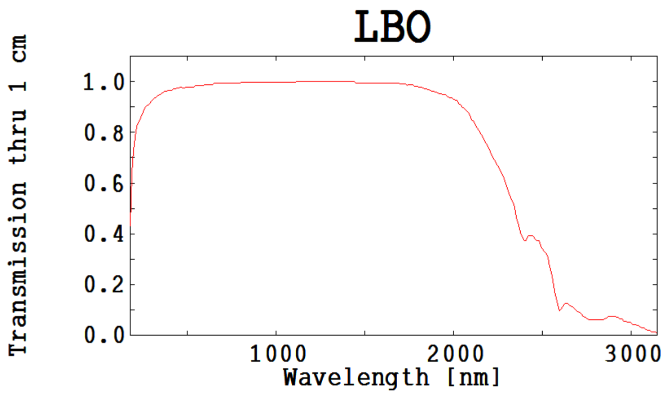


Figure 8: Transmittance over wavelength from LBO

LBO is, as explained in the previous chapter, a negative biaxial crystal. That means in fact, that the right crystal-plane for the simulation is the XY-plane. With this plane and the right tuning angle it is possible to achieve phase-matching. For selecting the correct angle Chi2D has a feature that can plot the tuning curves for different wavelengths, angles, types of polarizations and crystals as well as different non-linear processes.

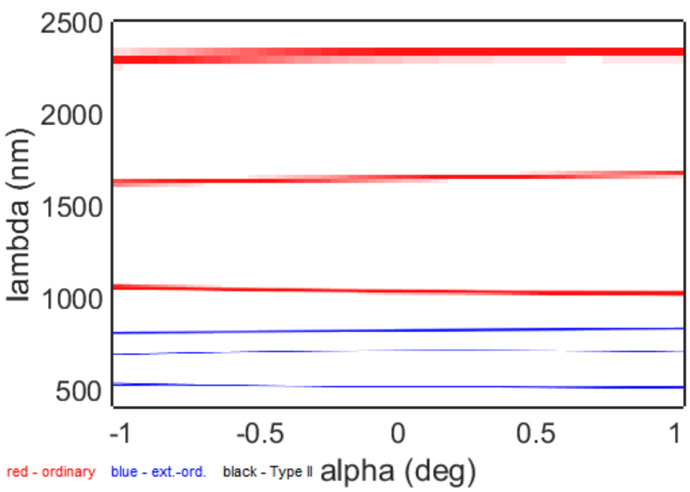
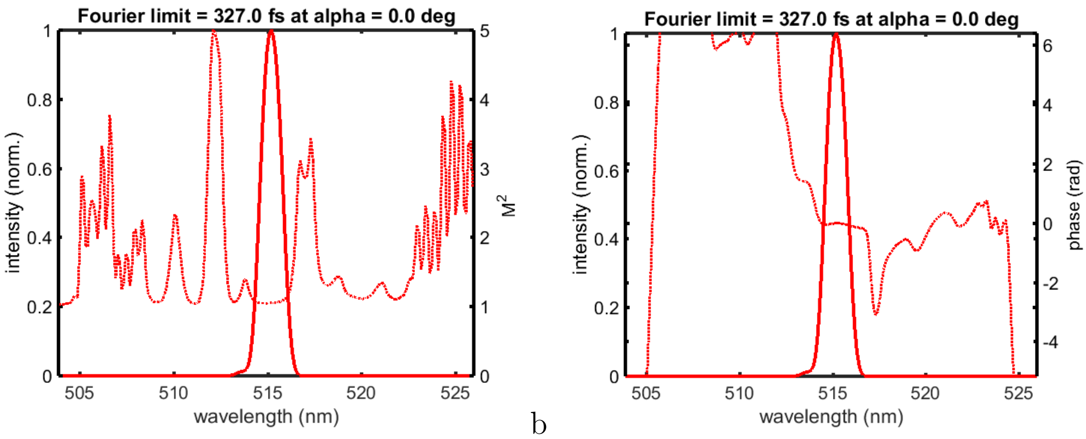


Figure 9: tuning curves for LBO

The plot in figure 9 shows tuning curves for the LBO crystal's XY-plane with an tuning angle of $\theta = 13,58°$. The blue graphs show phase matching for mixed polarizations, that means type two matching. The red graphs indicate phase matching for SHG and SFG with ordinary polarization. As one can see there is one line which fullfilles phase matching for the central wavelength of the laser puls at $\lambda = 1030nm$. So the tuning angle is setted right for an efficient SHG.
After setting up all the input parameters one has to decide about the framework conditions of the simulation. There the decision has to be make how high the resolution in time and space should be. For that it is always the goal to get the maximum out of the simulation and simultaneously minimising the calculation time. While some parameters are only for the function of the simulation some values have an enourmes importance for the practicle implementation in the lab later.
Chi2D has some powerfull data analysis tools, with whichsoever the conversion can be monitored.



a b

Figure 10: a: output spectrum with M^2 **b:** output spectrum with phase information

In this case the conversion efficiency was maximized with a, to $80\mu m$ down focused, beam. This is important for practical reasons later, because one has to bild optics that focus the beam exactly down to this waist. With this simulation it was possible to convert $4,9\mu J$ from the original $6\mu J$ into the second harmonic.

4 Experimental setup

Before setting up the crystal one has to reshape the output beam from the laser system. That however means that the characteristics of the beam must be knowen before, so that the right optical components could be choose to shape the beam as it is needed. Therefore the beam characteristics were measured with a camera and the divergence was calculated. The calculation brought out that the beam diverges different in sagittal and tangential plane. This leads then to different waists in those planes. The results of this calculation can be seen in the following table.

plane	waist [mm]	divergence [mrad]
sagittal	0.5396	0.61364
tangential	0.4664	0.70992

Table 1: beam characteristics by calculation

With this data it is possible to eveluate the right lenses to shape the beam correct. For this purpose a programm, called waistwatcher was used. The optical path of this beam can be seen in the next figure. There it can be seen that the foki from sagittal and tangential wave are at different positions z. This optical abberation is called astigmatism and can be correctet by turning one lens by an angle.

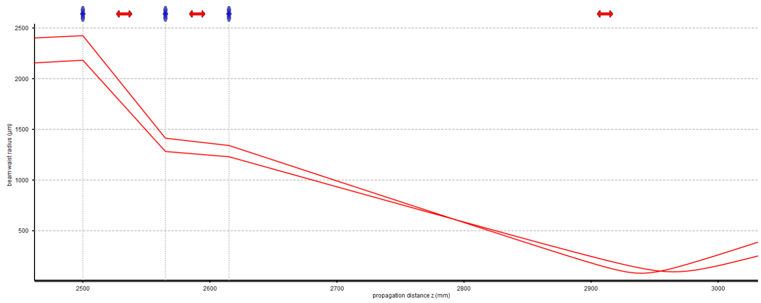


Figure 11: Optical path in telescope

The angle that was calculatet to bring both foki togther is at a value of 6°. Unfortunately, there happened a lot of small problems which then in sum lead to a serious time problem and it was not able to finish the project. That is to say, it was not able to take data from the SHG and compare this to the simluations. That is why there will no be disussion chapter in this report.

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